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# Self-humidifying membrane electrode assembly prepared by adding microcrystalline cellulose in anode catalyst layer as preserve moisture

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## ARTICLE INFO

### Article history:

Received 10 March 2014

Received in revised form

22 May 2014

Accepted 10 June 2014

Available online 7 July 2014

### Keywords:

Fuel cell

Membrane electrode assembly

Self-humidification

MCC

Hygroscopic agent

## ABSTRACT

A novel self-humidifying membrane electrode assemblies (MEAs) with the addition of microcrystalline cellulose (MCC) as a hygroscopic agent into anode catalyst layer was prepared to improve the performance of proton exchange membrane fuel cell (PEMFC) under low humidity conditions. The MEAs were characterized by SEM, contact angles and water uptake measurements. The MEAs with addition of MCC exhibit excellent self-humidifying single cell performance, the cell temperature for self-humidification running is up to 60 °C. As an optimized MEA with 4 wt.% MCC in its anode catalyst layer, its current density at 0.6 V could be up to 760 mA cm<sup>-2</sup> under 20% of relative humidity, and remains at 680 mA cm<sup>-2</sup> after 22 h long time continuous testing, the attenuation of the current density is only 10%. While the current density of the blank MEA without addition of MCC degraded sharply from 300 mA cm<sup>-2</sup> to 110 mA cm<sup>-2</sup>, the attenuation of the current density is high up to 70% within 2 h.

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## Introduction

Proton exchange membrane fuel cell (PEMFC) is considered as a promising green energy conversion technology in the future, due to its advantages, such as high energy conversion efficiency, low or zero polluting emissions and silent discharge [1–2]. However, its commercialization is obstructed by some of problems, low power density caused by the complex system and the water management are the two among these problems [3].

Membrane electrode assembly (MEA) is the essential component of PEMFC; it mainly consists of two catalyst layers

(cathode and anode) and an electrolyte membrane. Generally, the widely used perfluoride sulfuric acid membrane (such as Nafion membrane) should be kept wet in order to realize optimal proton conductivity, thus, we had to maintain the hydration of the Nafion® membrane during operation [4]. To ensure enough hydration, water is frequently introduced into the cell externally by humidifying the hydrogen and air prior to entering the cell. However, the additional humidifying devices bring a burden to fuel cell systems, complicate the system and lower the cell's energy efficiency. In response to this challenge, enormous attentions have been focused on the development of self-humidifying or non-humidifying

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<http://dx.doi.org/10.1016/j.ijhydene.2014.06.067>

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membrane electrode assemblies (MEA). One of the effective approaches is to modify the membrane by physical mixing inorganic oxide powder such as  $\text{SiO}_2$ ,  $\text{ZrO}_2$  and  $\text{TiO}_2$  with Nafion solution or through the sol gel reactions to improve the water retention of the membrane [5–8]. The inorganic oxides act as water absorbers can improve proton conductivity by keeping the membrane in a well-hydrated condition under low humidity or dry conditions. Some authors [9–11] have incorporated Pt nanoparticles along with oxides particles into the membrane to generate water and adsorb water molecules produced from Pt particles to maintain the membrane with self-humidifying ability at the elevated temperature and low humidity operation of PEMFC.

Another method is to prepare self-humidifying MEA or optimize the MEA's structure by adding inorganic oxide particles into the catalyst layer [12–18]. Jung et al. [12] directly dispersed hydrophilic  $\text{SiO}_2$  particles into the anode catalyst layer to control the wettability and performance of MEA at low-humidity conditions, and the low-humidity performance of the MEA with 40 wt.%  $\text{SiO}_2$  to the anode was improved significantly. Su et al. [13] fabricated a Pt– $\text{SiO}_2$ /C self-humidifying catalyst via organic colloid method. When this catalyst was used in anode CL, At a cell temperature of 50 °C and 28% of RH for both the anode and cathode, the current density at 0.6 V remained at 0.65 A  $\text{cm}^{-2}$  without any degradation for 20 h. Kannan et al. [16,17] prepared self-humidifying MEAs by adding inorganic oxides (such as  $\text{SiO}_2$ ,  $\text{TiO}_2$  and  $\text{Al}_2\text{O}_3$ ) on GDL. The MEAs with a thin layer of oxide showed a high performance even at low humidifying conditions.

Some researchers have prepared self-humidifying MEA by adding hydrophilic organic polymer as hygroscopic agent [19–23]. Cindrella et al. [19] designed a MEA by incorporating a thin layer of doped polyaniline (PANI) between the catalyst layer and the micro-porous layer (MPL) to improve the low relative humidity (RH) performance of PEMFCs. Kitahara et al. [21] developed a new hydrophilic and hydrophobic double MPL coated GDL to achieve further enhancement of performance under non-humidification at the cathode. Zhang et al. [23] proposed sulfonated poly(fluorenyl ether ketone)s (SPFEK) membranes by using organic polymer hydroxypropyl methyl cellulose (HPMC) as dispersant. The results demonstrated HPMC is an efficient dispersant for the organic/inorganic hybrid membrane used for PEMFC at high temperature and low relative humidity (RH). However, current achievements in self-humidification MEA still have a big gap to match the requirements for the practically application.

Microcrystalline cellulose (MCC), which has insolubility, chemical inactivity, absence of toxicity and great hygroscopicity, is widely used in pharmaceutical, cosmetic and food industries [24]. Nevertheless, to our knowledge, there is no report of the application of organic polymer microcrystalline cellulose as hygroscopic agent in the composite catalyst layer in PEMFCs. As one of the continuous attempts to obtain high performance self-humidification MEAs, we attempted to prepare self-humidifying MEAs by introducing microcrystalline cellulose (MCC) in the anode catalyst layer. Excitingly, the MEAs exhibit excellent self-humidifying performance at low humidity of 20% RH conditions and the cell temperature for stable low humidity operation is high up to 60 °C.

## Experiment

### Preparation of MEAs

The MEAs with an active area of 5  $\text{cm}^2$  were fabricated using a direct catalyst spraying technique developed in our laboratories previously [25]. Firstly, Nafion 212 membranes were pretreated with the procedures reported previously by our research group. Anode catalyst inks were prepared by mixing a commercial Johnson Matthey Hispec-4100 Pt/C catalyst (40 wt.%) (Denoted as JM Pt/C), microcrystalline cellulose powder, isopropanol and 5 wt.% Nafion solution (DuPont, USA). The weight percentages of MCC to the amount of catalyst, dry Nafion and dry MCC are 2 wt.%, 4 wt.%, 6 wt.%, and 8 wt.% respectively. The cathode catalyst ink was prepared by the same method but without MCC. The Pt loadings at the anode and cathode were 0.1  $\text{mg cm}^{-2}$  and 0.2  $\text{mg cm}^{-2}$  respectively, and the dry Nafion content in the anode and cathode catalyst layers was adjusted to 25 wt.%. Then, the inks were sprayed on both sides of Nafion 212 membrane (DuPont, USA) to form the catalyst layers. The MEAs with MCC content of 2 wt.%, 4 wt.%, 6 wt.% and 8 wt.% in anode were denoted in abbreviated form as MEA-2, MEA-4, MEA-6, MEA-8, one MEA prepared using JM Pt/C catalyst both at the anode and cathode was denoted as MEA-0. Morphology of the MEA-0 and MEA-4 was observed by Hitachi S-3700 scanning electron microscope (SEM).

### Measurement of contact angle of anode catalyst layer

The sessile drop method is the most common used means of measuring the contact angle of a liquid on a solid surface. In this technique, a droplet of water was set on the surface and the contact angle was measured by fitting a tangent to the three-phase point where the liquid surface touched the solid surface. To characterize the wetting property of anode catalyst layer of MEAs, the contact angle of a water drop on the catalyst layer was determined experimentally by direct image analysis of the shape of the drop at room temperature. The measurements were performed with an OCA 40 Video-Based Automatic Contact Angle Meter (Dataphysics, Germany).

### Measurement of water uptake

Water uptake of the anode catalyst layer was calculated by weighing the weight change of wet and dry samples of anode catalyst layer [22]. The samples were made by spraying the above anode catalyst inks onto one side waterproof carbon paper with an area of 6.25  $\text{cm}^2$ . Followed by drying in a vacuum oven at 80 °C for 4 h. The water uptake of composite catalyst layer is referring to the liquid water reserved in the sample and is expressed as:

$$\Delta W(\text{wt.}\%) = \frac{W_{\text{water}}}{W_{\text{CL}}} \times 100\%$$

Where  $W_{\text{water}}$  is the weight of liquid water reserved in each sample, and  $W_{\text{CL}}$  is the weight of composite catalyst, which is obtained by subtracting the weight of the carbon paper from the total. In order to measure the  $W_{\text{water}}$ , the sample was first dried in a vacuum oven at 105 °C for 4 h to obtain the dry

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